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13. ABSTRACT (Maximum 200 words) This report results from a contract tasking University of Leeds as follows: The contractor will investigate the development of Pr3+-doped fluoroindate and fluoroaluminate host glasses to determine if the chemistry of their composition can be adjusted so that useful gain at 589 nm can be achieved. 8 glass samples will be delivered to AFRL for evaluation. <div style="text-align: center; font-size: 2em; font-weight: bold;">20010126 044</div>				
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Pr³⁺ -doped fluoride glass for a 589 nm fibre laser

**University of Leeds
Principal investigator: Dr Animesh Jha**

**Final Report
1 May 1999 - 20 December 1999**

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see also our previous Reports on this project

AQ F01-04-0744

See also the enclosed copy of our paper submitted to Journal of Luminescence

1. INTRODUCTION

We have undertaken to investigate a range of Pr^{3+} -doped fluoride glasses with the aim of identifying a suitable glass host for a 589 nm laser. Pr^{3+} -doped fluorozirconate ZBLAN glass is known to lase at 601-618 nm, but lasing at 589 nm cannot be achieved in this host.

In preliminary experiments with different Pr^{3+} -doped fluoride glasses we observed that the emission spectrum of Pr^{3+} depends strongly on the host glass. Figure 1 in the attached paper shows the energy levels of Pr^{3+} . When excited with blue and violet lines of Ar^+ laser, Pr^{3+} emits red fluorescence consisting of two peaks (Figures 3 and 4 in the paper), $^3\text{P}_0 \rightarrow ^3\text{H}_6$ at 603 nm and $^3\text{P}_0 \rightarrow ^3\text{F}_2$ at 635 nm, which both originate from the $^3\text{P}_0$ level. The short-wavelength shoulder of the emission peak at 603 nm contains a small contribution from the $^3\text{P}_1 \rightarrow ^3\text{F}_2$ transition. As seen in Figure 1, the $^3\text{P}_0$ and $^3\text{P}_1$ levels lie sufficiently close together ($\sim 500 \text{ cm}^{-1}$) to be thermalized; at room temperature the population of the $^3\text{P}_1$ level is around 6%. It is evident from the emission spectra (Figures 4 and 3 in the paper) that the $^3\text{P}_1 \rightarrow ^3\text{F}_2$ fluorescence peak lies at approximately 585 nm; a similar value is derived from the absorption spectrum shown in Figure 2. Therefore $^3\text{P}_1 \rightarrow ^3\text{F}_2$ fluorescence can provide a major contribution to the 589 nm laser. By heating the glass, the $^3\text{P}_1$ level can be populated and its emission greatly increased.

As demonstrated in the paper, the population of the $^3\text{P}_1$ level N_1 is determined by the glass temperature T and the $^3\text{P}_1$ - $^3\text{P}_0$ energy gap ΔE according to the Maxwell-Boltzmann distribution:

$$N_1 = e^{-\Delta E/kT}$$

Consequently, the population N_1 at a given temperature can be increased by reducing the energy gap ΔE . In our investigations we have found that ΔE varies significantly in different glass hosts (see Figure 5 and Table 1 of the paper). For the purposes of laser design, it is obviously advantageous to operate the device at lower temperatures, therefore the last phase of our project was devoted to designing a glass host which would minimise ΔE .

2. EXPERIMENTAL

Luminescence from the $^3\text{P}_0$ level was excited by blue and violet lines of an Ar^+ laser, and was clearly observed as a bright pink track in the sample along the pump beam. The spectra were measured by a scanning spectrometer equipped with a Si-photodetector. The instrument was calibrated to produce true amplitudes across the recorded spectrum. The samples were heated on a stage and the temperature was monitored by a thermocouple.

3. RESULTS

Our aim was to design an improved glass host for the Pr^{3+} -doped 589 nm laser. The glass must be highly stable, and the energy gap ΔE must be substantially reduced compared with the fluoroaluminate glass described in our paper. We limited our investigations to modifying fluoroaluminate glass compositions, since these glasses have high glass transitions temperatures, good stability, and have demonstrated lower ΔE values than indium fluorides and zirconium fluorides (see our previous Intermediate Report).

We have found the ΔE is reduced when Na compounds are removed from the glass. Phosphates in the form of metaphosphates also have a slight beneficial effect on ΔE , while greatly improving the glass stability and resistance to devitrification. Heavy fluoroaluminate compositions, containing YF_3 and ZrF_4 , performed better than lighter ones, both spectroscopically and as more stable glasses.

The optimised glass composition is ALF5994:



Figure 1 shows the 585nm/604nm intensity ratio as a function of temperature in three glasses: ALF5994, ALF599 (described in the paper), and ZBLAN. The reason for plotting the 585nm/604nm ratio and the calculation of the fitted curves are described in our paper. It is seen that in ALF5994 this ratio rises more steeply. The calculated 3P_1 - 3P_0 energy gap ΔE is 470 cm^{-1} in ALF5994 and 515 cm^{-1} in ALF599.

Figure 2 shows the 589nm/604nm intensity ratio as a function of temperature in ALF5994 and ALF599. Again it is seen that at a given temperature ALF5994 exhibits a larger relative intensity at 589 nm. Emission intensity of 50% (3 dB) at 589 nm is achieved at 270°C in ALF5994 compared with 320°C in ALF599. These results are summarised in the table below.

Glass	ALF5994	ALF599
ΔE (cm^{-1})	470 \pm 10	515 \pm 10
Temperature where 589nm/604nm is at 50%	270°C	320°C

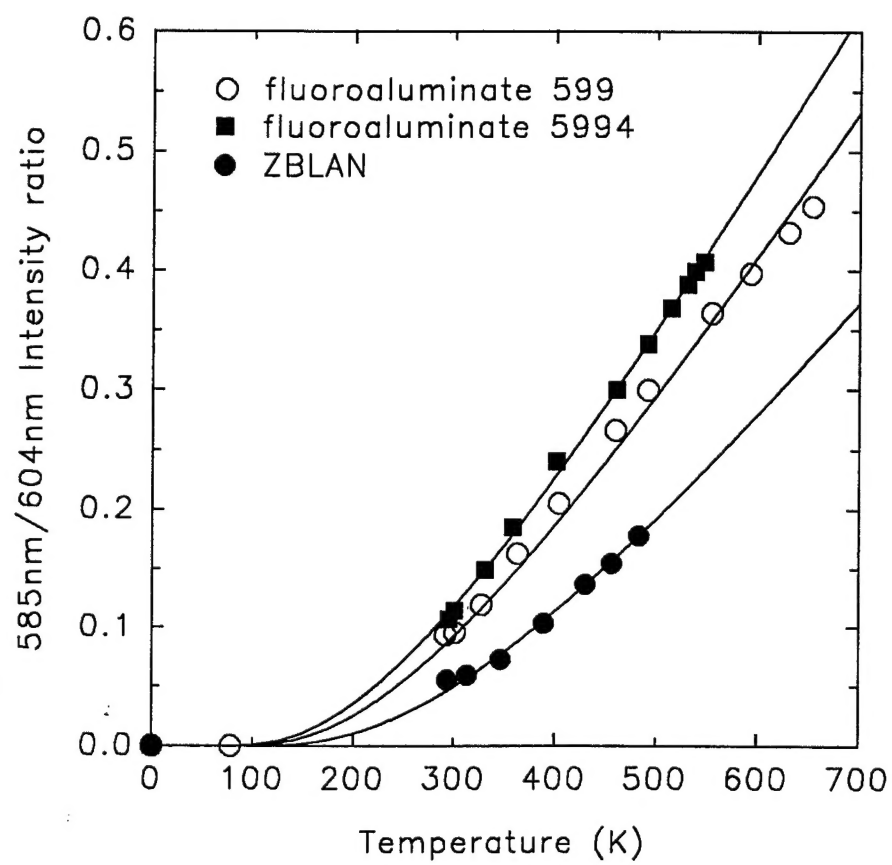
Figure 3 shows a DSC (dynamic scanning calorimeter) scan of glass ALF5994, recorded isochronally at 10°C/min. It is seen that the glass softening point T_g is at 410°C and the onset of crystallisation T_x is at 520°C. The T_x - T_g gap is 110°C, and the onset of crystallisation is relatively shallow, with peak crystallisation T_p being at 565°C. These features confirm the good stability of ALF5994 glass, which was also observed during the fabrication process.

4. CONCLUSIONS

We have shown that fluoroluminate glass ALF5994 is a possible host for a Pr^{3+} -doped laser at 589 nm. Emission at 589 nm is obtained by heating the glass to thermally populate the 3P_1 level. It is estimated that the laser will operate at temperatures of 250-280°C. This is > 130°C below the softening point of the glass. Our experience shows that the glass is not damaged by being maintained for a long period at this temperature. DSC results confirm that the glass is highly stable, and that it is not liable to devitrify or deform under these conditions. Therefore a 589 nm laser device utilising Pr^{3+} -doped ALF5994 glass appears to be possible.

5. FABRICATION OF GLASS RODS

Glass rods of Pr^{3+} -doped ALF5994 glass are at present being fabricated for fibre drawing and lasing experiments. We have identified an impurity contamination problem, and are awaiting the shipment of higher purity materials. Glass rods fabricated from high purity fluoride powders will be supplied to EOARD towards the middle of January 2000.



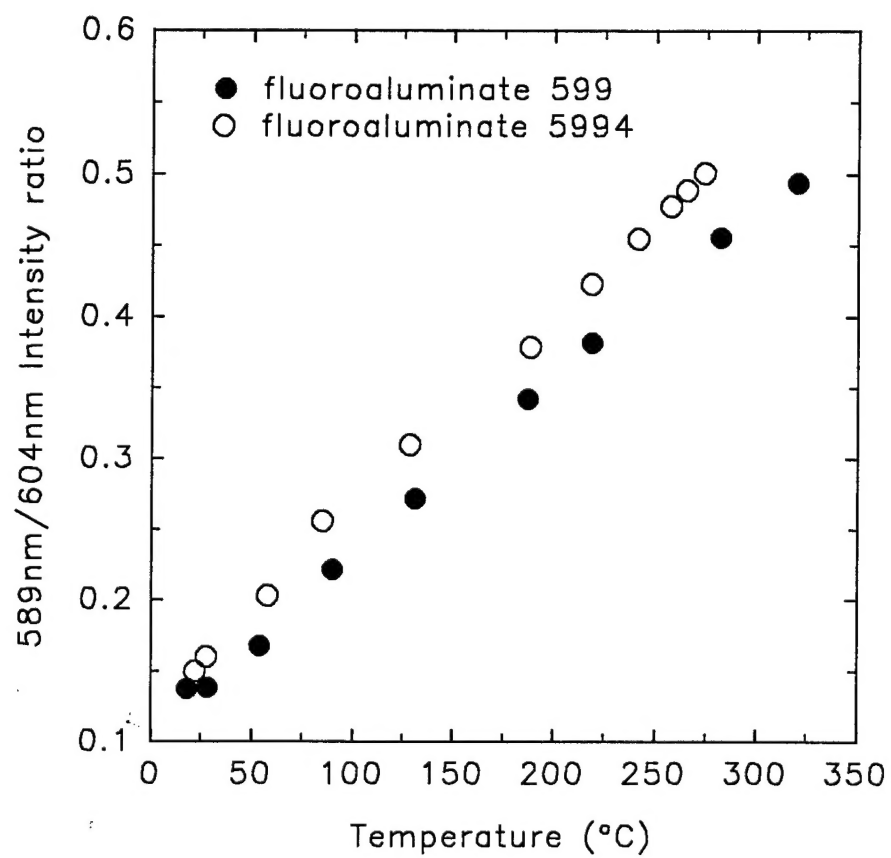
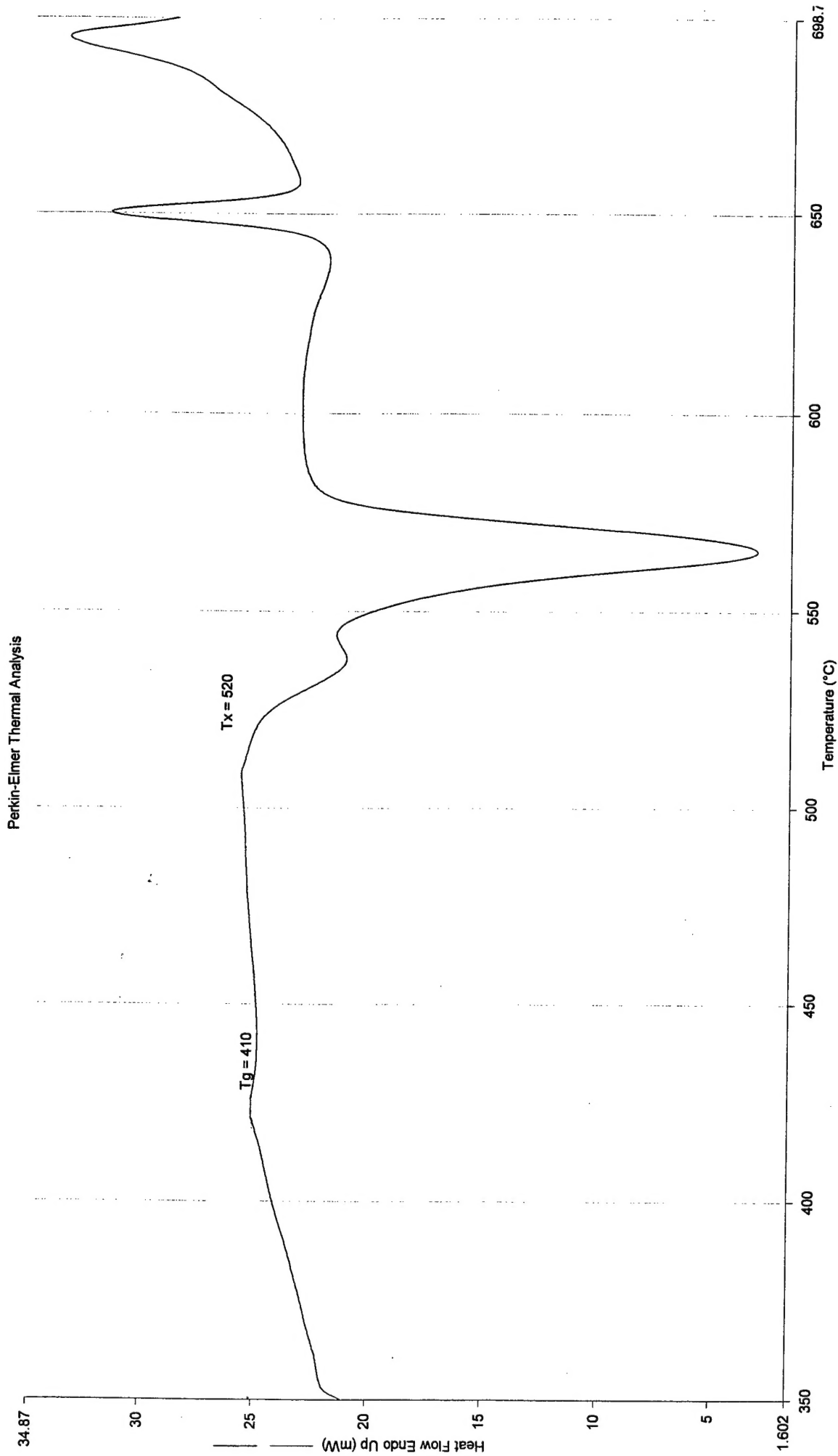


FIG 2

Filename: C:\PE\Pyri...alf5994@991210155839.ddd
Operator ID: C.Batchelor
Sample ID: alf5994
Sample Weight: 14.400 mg
Comment: Baseline subtracted



1) Hold for 1.0 min at 350.00°C

2) Heat from 350.00°C to 700.00°C at 10.00°C/min

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